LEAD SOIL TREND ANALYSIS
THROUGH OCTOBER, 2005
EVALUATION BY INDIVIDUAL QUADRANT
Herculaneum Lead Smelter Site
Herculaneum, Missouri

Tetra Tech EM Inc. (Tetra Tech) was tasked by the U.S. Environmental Protection Agency (EPA) Region 7 Enforcement/Fund Lead Removal program to conduct a trend analysis of soil lead concentrations at selected locations within Herculaneum, Missouri (City). Specifically, the Tetra Tech Superfund Technical Assessment and Response Team (START) 2 was requested to review and analyze data that would enable EPA to determine if soil lead concentrations were increasing over time at a variety of locations within the City. Two tasks were identified: 1) perform a trend analysis for individual quadrants within each yard using the most current sampling data, and 2) estimate the range of monthly increase in lead concentrations for properties grouped into three categories based on distance from the smelter (less than or equal to 025 mile, 0.25 to 0.50 miles, and 0.50 to 0.75 miles). The assessment was conducted under the authority of the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 and the Superfund Amendments and Reauthorization Act of 1986. The project was assigned under START Contract No. 68-S7-01-41, Task Order No. 0027.

Tetra Tech focused its analysis on one data set called "Recontamination." This data set includes results from a number of residential properties. The data were collected from four different quadrants at each property, and additional data for several properties came from samples collected in driveway areas outside the quadrants. Lead concentrations were estimated at each location at approximately monthly intervals from the time removal activities were completed until October 2005 (sampling round 22). Due to the sequence of removal activities, not all properties underwent the same number of sampling events; the number of events ranged from 5 to 16 events per quadrant for individual properties. At many locations, some intervals within the series were omitted because of weather or access restrictions. The lead concentrations were determined by use of a portable X-ray fluorescence (XRF) instrument. Samples were collected and analyzed in accordance with the quality assurance project plan (QAPP) dated September 11, 2001.

This document presents the methods used to evaluate changes in soil lead concentrations following the removal activities, and the results of this analysis.

Methods

Trend tests were conducted for each property using data collected from round 7 (August 2002) through round 22 (October 2005). The non-parametric Mann-Kendall test was used to evaluate temporal trends for each sampled quadrant at the individual properties. The Mann-Kendall test is a widely used statistical test for detecting monotonic trends (that is, trends that are either increasing or decreasing) in time-series of data (Gilbert 1987; Helsel and Hirsch 1992; Gibbons 1994). Because the Mann-Kendall test uses only the relative magnitude of the data rather than their measured values, it has a number of desirable properties: the data need not be normally distributed; and the test is not significantly affected by outliers, missing data, or censored data. Censored data are treated in the Mann-Kendall test by setting all non-detect values to a concentration slightly below the minimum detected concentration. It should be noted that a minimum of four sampling events are required to perform this test, so properties with fewer than four rounds of sampling were not evaluated. Properties which were not sampled during round 22 were also excluded from the trend analysis.

For all properties where at least one quadrant showed a significant increasing trend based on the Mann-Kendall test, regression analysis was performed to estimate the monthly increase in lead concentration. This analysis was performed to provide rough estimates of the range of potential increase in lead concentrations for properties grouped according to distance from the smelter. Three distance categories were evaluated: less than or equal to 0.25 miles, 0.25 to 0.50 miles, and 0.50 to 0.75 miles. Because the purpose of this analysis was to only provide rough estimates of the rate of change in lead concentration, regression was performed on the data in original units (i.e., untransformed data). It should be noted that certain evaluation methods and diagnostic tools that are commonly used in linear regression analysis (e.g., evaluation of different transformations of the data, verification of model assumptions, and evaluation of outliers) were not used in this analysis.

For quadrants with detected data only, ordinary least squares (OLS) linear regression analysis was used. For quadrants with one or more censored (nondetect) measurements, a censored maximum likelihood estimation (MLE) approach was used, following Helsel (2005). Censored MLE methods are increasingly being used in environmental assessment work, given the increased speed of modern personal computers and the enhanced capabilities that have been added into many commercial statistical software packages. As described in Helsel (2005), MLE regression techniques can be implemented using commercial software with capabilities for performing parametric survival analysis on interval-censored data. It should be noted that MLE regression for left-censored data is also referred to as "Tobit analysis" in the technical literature. MLE methods recognize each censored datum as an interval, bounded by zero at the lower limit and the detection or reporting limit at the upper limit. Application of OLS regression with censored data is contraindicated, as it requires substitution of an assumed value (typically zero, the detection limit, or one half the detection limit) for each censored datum, resulting in biased estimates for the regression parameters.

Results

Temporal trends in lead concentrations for 16 properties are summarized in Table 1 and Figure 1. The trend analysis identified 15 out of 16 properties where at least one quadrant showed a statistically significant increasing trend. No statistically significant decreasing trends were identified for any properties. Six properties had increasing lead concentrations in all four quadrants: House numbers 5, 9, 18, 19, 20, and 22. Two properties had increasing lead concentrations in three of four quadrants: House Numbers 16 and 24. Five properties had increasing lead concentrations in two of four quadrants: House numbers 3, 6, 7, 76 (only two quadrants evaluated), and 102. House number 101 had only one quadrant with an increasing trend in lead concentration. Only one property, House number 15, showed no statistically significant trend in lead concentrations in any quadrant. All trend results are depicted graphically in Figure 1. Open symbols are used in Figure 1 to represent censored (nondetect) data, and solid symbols represent detected data.

Trend results reported for soil lead concentrations through sampling round 22 were similar to those reported during the last quarterly period, with the following exceptions. Six quadrants from six properties that did not show a significant trend in lead concentration from rounds 7 through 21, now show a statistically significant increase in lead concentration with the addition of the data from round 22. The properties include House numbers 6 (quadrant 1), 18 (quadrant 2), 19 (quadrant 3), 24 (quadrant 3), 76 (quadrant 2), and 102 (quadrant 4). Three quadrants from three properties that showed a significant trend in lead concentration from rounds 7 through 21, now show no statistically significant increase in lead concentration with the addition of the data from round 22. The properties include House numbers 7 (quadrant 3), 101 (quadrant 3), and 102 (quadrant 1).

The results of OLS and MLE regression analysis performed on properties that showed a significant increasing trend in lead concentration in at least one quadrant are provided in Table 2. The slope, intercept, standard error of the slope, and two-sided 95 percent confidence intervals for the slope estimates were calculated for 43 quadrants within 14 properties. Ranges for the monthly rates of increase in lead were 1.17 to 10.92 milligrams (mg)/month, 1.38 to 6.75 mg/month, and 0.95 to 9.90 mg/month, respectively, for properties located less than or equal to 0.25 miles, 0.25 to 0.50 miles, and 0.50 to 0.75 miles from the smelter. The upper 95 percent confidence limit (UCL) for the monthly rate of increase was also evaluated to estimate maximum potential rates of increase. Because of the variability in the individual estimates, the 50^{th}, 75^{th}, and 90^{th} percentiles of the distribution of the individual UCLs within each distance category are also reported in Table 2. The 75^{th} and 90^{th} (in parentheses) percentile values for the monthly rate of increase for the properties grouped according to increasing distance from the smelter are 10.17 (17.53), 7.26 (12.59), and 3.94 (14.27) mg/month. It should be cautioned that these are considered rough estimates only, as no attempt was made to evaluate the validity of the regression model assumptions, or the uncertainty associated with the predicted rates of increase.

References:

Gibbons, R. D. 1994. *Statistical Methods for Groundwater Monitoring*. John Wiley & Sons, Inc. New York, New York.

Gilbert, R. O. 1987. *Statistical Methods in Environmental Pollution Monitoring*. John Wiley & Sons, Inc. New York, New York.

Helsel, D. 2005. *Nondetects and Data Analysis: Statistics for Censored Environmental Data*. John Wiley & Sons, Inc., New York, NY. 250 p.

Helsel, D. R. and R. M. Hirsh. 1992. *Statistical Methods in Water Resources*. Elsevier. New York, New York.

TABLE 1
RESULTS OF STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATION
INDIVIDUAL QUADRANTS FOR SAMPLING ROUNDS 7 THROUGH 22
HERCULANEUM LEAD SMELTER SITE - HERCULANEUM, MISSOURI

Distance From Smelter[1]	House Number	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event		Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
					First	Last				
0.10	76	Q1	9	9	10/30/2003	10/03/2005	24	0.006	Yes	**Increasing**
		Q2	9	9	10/30/2003	10/03/2005	18	0.038	Yes	**Increasing**
0.20	20	Q1	15	15	08/26/2002	10/04/2005	63	0.002	Yes	**Increasing**
		Q2	15	15	08/26/2002	10/04/2005	61	0.002	Yes	**Increasing**
		Q3	15	15	08/26/2002	10/04/2005	77	0.000	Yes	**Increasing**
		Q4	15	15	08/26/2002	10/04/2005	67	0.001	Yes	**Increasing**
	101	Q1	8	8	12/22/2003	10/04/2005	8	0.199	No	N/A
		Q2	8	7	12/22/2003	10/04/2005	12	0.089	No	N/A
		Q3	8	8	12/22/2003	10/04/2005	12	0.089	No	N/A
		Q4	8	8	12/22/2003	10/04/2005	16	0.031	Yes	**Increasing**
	102	Q1	8	8	12/22/2003	10/04/2005	14	0.054	No	N/A
		Q2	8	8	12/22/2003	10/04/2005	0	0.548	No	N/A
		Q3	8	8	12/22/2003	10/04/2005	18	0.016	Yes	**Increasing**
		Q4	8	8	12/22/2003	10/04/2005	18	0.016	Yes	**Increasing**
0.25	5	Q1	15	12	08/26/2002	10/03/2005	77	0.000	Yes	**Increasing**
		Q2	15	14	08/26/2002	10/03/2005	77	0.000	Yes	**Increasing**
		Q3	15	15	08/26/2002	10/03/2005	72	0.000	Yes	**Increasing**
		Q4	15	15	08/26/2002	10/03/2005	63	0.002	Yes	**Increasing**
	6	Q1	15	15	08/23/2002	10/04/2005	37	0.041	Yes	**Increasing**
		Q2	15	15	08/23/2002	10/04/2005	63	0.002	Yes	**Increasing**
		Q3	15	15	08/23/2002	10/04/2005	14	0.162	No	N/A
		Q4	15	15	08/23/2002	10/04/2005	31	0.066	No	N/A
	22	Q1	14	14	08/26/2002	10/04/2005	37	0.029	Yes	**Increasing**
		Q2	14	14	08/26/2002	10/04/2005	39	0.023	Yes	**Increasing**
		Q3	14	14	08/26/2002	10/04/2005	48	0.007	Yes	**Increasing**
		Q4	14	14	08/26/2002	10/04/2005	47	0.008	Yes	**Increasing**
	24	Q1	12	12	11/07/2002	10/04/2005	20	0.085	No	N/A
		Q2	12	12	11/07/2002	10/04/2005	44	0.003	Yes	**Increasing**
		Q3	12	12	11/07/2002	10/04/2005	30	0.028	Yes	**Increasing**
		Q4	12	11	11/07/2002	10/04/2005	43	0.003	Yes	**Increasing**
0.40	21	Q1	11	9	08/23/2002	10/03/2005	24	0.040	Yes	**Increasing**
		Q2	11	11	08/23/2002	10/03/2005	31	0.013	Yes	**Increasing**
		Q3	11	11	08/23/2002	10/03/2005	34	0.007	Yes	**Increasing**
		Q4	11	11	08/23/2002	10/03/2005	45	0.001	Yes	**Increasing**

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TABLE 1
RESULTS OF STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATION
INDIVIDUAL QUADRANTS FOR SAMPLING ROUNDS 7 THROUGH 22
HERCULANEUM LEAD SMELTER SITE - HERCULANEUM, MISSOURI

Distance From Smelter[1]	House Number	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event First	Sampling Event Last	Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
0.50	15	Q1	5	4	09/16/2002	10/04/2005	2	0.408	No	N/A
		Q2	5	5	09/16/2002	10/04/2005	5	0.180	No	N/A
		Q3	5	4	09/16/2002	10/04/2005	1	0.500	No	N/A
		Q4	5	4	09/16/2002	10/04/2005	6	0.117	No	N/A
	16	Q1	13	9	09/16/2002	10/04/2005	20	0.100	No	N/A
		Q2	13	7	09/16/2002	10/04/2005	57	0.000	**Yes**	**Increasing**
		Q3	13	7	09/16/2002	10/04/2005	35	0.018	**Yes**	**Increasing**
		Q4	13	9	09/16/2002	10/04/2005	56	0.001	**Yes**	**Increasing**
	19	Q1	15	14	08/22/2002	10/04/2005	46	0.017	**Yes**	**Increasing**
		Q2	15	12	08/22/2002	10/04/2005	38	0.037	**Yes**	**Increasing**
		Q3	15	12	08/22/2002	10/04/2005	40	0.030	**Yes**	**Increasing**
		Q4	15	14	08/22/2002	10/04/2005	55	0.006	**Yes**	**Increasing**
0.54	9	Q1	15	15	08/22/2002	10/04/2005	54	0.006	**Yes**	**Increasing**
		Q2	15	15	08/22/2002	10/04/2005	52	0.008	**Yes**	**Increasing**
		Q3	15	15	08/22/2002	10/04/2005	57	0.004	**Yes**	**Increasing**
		Q4	15	14	08/22/2002	10/04/2005	54	0.006	**Yes**	**Increasing**
0.60	18	Q1	16	16	08/23/2002	10/03/2005	54	0.012	**Yes**	**Increasing**
		Q2	16	15	08/23/2002	10/03/2005	44	0.031	**Yes**	**Increasing**
		Q3	16	16	08/23/2002	10/03/2005	57	0.008	**Yes**	**Increasing**
		Q4	16	16	08/23/2002	10/03/2005	60	0.006	**Yes**	**Increasing**
0.75	3	Q1	16	13	08/23/2002	10/03/2005	19	0.143	No	N/A
		Q2	16	14	08/23/2002	10/03/2005	61	0.005	**Yes**	**Increasing**
		Q3	16	15	08/23/2002	10/03/2005	26	0.106	No	N/A
		Q4	16	15	08/23/2002	10/03/2005	67	0.002	**Yes**	**Increasing**
0.80	7	Q1	16	16	08/23/2002	10/04/2005	37	0.053	No	N/A
		Q2	16	14	08/23/2002	10/04/2005	69	0.002	**Yes**	**Increasing**
		Q3	16	12	08/23/2002	10/04/2005	36	0.056	No	N/A
		Q4	16	12	08/23/2002	10/04/2005	72	0.001	**Yes**	**Increasing**

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Notes:

[1] Properties are ordered as a function of increasing distance from the smelter.

[2] Trend tests were not conducted for properties with fewer than four rounds of sampling, or for properties that were not sampled during round 22.

[3] All censored (nondetect) measurements were set equal to a concentration slightly lower than the minimum detected value

[4] Monotonic trends are significant for probabilities less than or equal to 0.05; significant negative values for the Mann-Kendall test statistic indicate that trends are decreasing; and significant positive values for the Mann-Kendall test statistic indicate that trends are increasing.

NA No significant trend identified.

TABLE 2
RESULTS OF LINEAR REGRESSION ANALYSIS FOR ALL QUADRANTS SHOWING A SIGNIFICANT INCREASING MANN-KENDALL TREND TEST RESULT

Distance From Smelter (Miles)	House Number	Quadrant	Regression Coefficients for Days Versus Concentration			Monthly Increase (mg/kg-month)	95 Percent Confidence Limits for Monthly Increase in Lead Concentrations		Percentiles for the Distribution of Estimated UCLs within Each Distance Group		
			Intercept	Slope	S.E. (Slope)		LCL	UCL	50	75	90
Less than or Equal to 0.25	76	Q1	8.24	0.21	0.05	6.34	2.61	10.07	5.81	10.17	17.53
	76	Q2	72.95	0.12	0.12	3.61	-4.79	12.01			
	20	Q1	91.48	0.14	0.04	4.29	1.98	6.61			
	20	Q2	51.15	0.28	0.06	8.42	4.78	12.05			
	20	Q3	100.62	0.20	0.04	6.07	3.66	8.48			
	20	Q4	90.81	0.25	0.05	7.49	4.52	10.47			
	101	Q4	-10.73	0.14	0.05	4.24	0.74	7.73			
	102	Q3	32.30	0.36	0.13	10.92	1.46	20.37			
	102	Q4	50.16	0.36	0.12	10.91	1.95	19.88			
	5	Q1	26.79	0.12	0.02	3.70	2.36	5.05			
	5	Q2	40.71	0.09	0.01	2.79	1.86	3.72			
	5	Q3	64.02	0.12	0.02	3.47	2.02	4.92			
	5	Q4	70.22	0.15	0.04	4.53	2.19	6.86			
	6	Q1	119.86	0.05	0.04	1.64	-1.11	4.38			
	6	Q2	79.36	0.13	0.03	3.80	1.76	5.84			
	22	Q1	94.11	0.08	0.03	2.26	0.53	3.99			
	22	Q2	182.38	0.11	0.04	3.41	1.05	5.77			
	22	Q3	73.62	0.08	0.03	2.49	0.67	4.32			
	22	Q4	70.70	0.10	0.03	2.89	0.85	4.93			
	24	Q2	40.64	0.11	0.02	3.31	1.78	4.83			
	24	Q3	63.54	0.04	0.01	1.17	0.37	1.97			
	24	Q4	48.47	0.09	0.02	2.84	1.41	4.28			
0.25 to 0.50	21	Q1	55.78	0.23	0.10	6.75	-0.23	13.74	4.83	7.26	12.59
	21	Q2	78.56	0.20	0.03	5.98	3.98	7.98			
	21	Q3	49.81	0.13	0.03	4.04	2.22	5.86			
	21	Q4	49.27	0.14	0.01	4.15	3.46	4.83			
	16	Q2	14.50	0.19	0.02	5.70	4.51	6.89			
	16	Q3	60.09	0.05	0.02	1.38	0.08	2.69			
	16	Q4	42.43	0.18	0.03	5.53	3.80	7.26			
	19	Q1	52.98	0.05	0.02	1.47	0.48	2.46			
	19	Q2	47.82	0.05	0.01	1.46	0.54	2.38			
	19	Q3	38.80	0.05	0.02	1.43	-0.01	2.87			
	19	Q4	54.14	0.07	0.02	2.16	0.86	3.46			
0.50 to 0.75	9	Q1	67.59	0.04	0.02	1.34	0.29	2.39	2.86	3.94	14.27
	9	Q2	65.51	0.08	0.02	2.37	0.97	3.76			
	9	Q3	69.23	0.33	0.08	9.90	4.45	15.36			
	9	Q4	88.02	0.10	0.02	2.86	1.26	4.47			
	18	Q1	68.53	0.06	0.02	1.86	0.75	2.97			
	18	Q2	46.25	0.07	0.02	2.19	1.18	3.20			
	18	Q3	69.94	0.03	0.01	0.95	0.31	1.59			
	18	Q4	58.79	0.06	0.01	1.69	0.88	2.49			
	3	Q2	47.74	0.06	0.02	1.68	0.63	2.74			
	3	Q4	44.69	0.05	0.01	1.36	0.64	2.08			

Notes:

LCL Lower confidence limit
MLE Maximum likelihood estimation
ND Nondetect
OLS Ordinary least squares
S.E. Standard error of estimate
UCL Upper confidence limit

OLS regression was used for cases where all results were detected. Censored MLE regression was used in all cases where one or more measurements were reported as below the detection limit (that is, "ND") following Helsel (2005). All analyses were performed on the data in original units.

Helsel, D. 2005. *Nondetects and Data Analysis: Statistics for Censored Environmental Data*. John Wiley & Sons, Inc., New York, NY. 250 p.